

N 70 26979

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EFFECT OF ADDING LITHIUM NITRIDE, HAFNIUM, TANTALUM,
AND TUNGSTEN TO A FAST-SPECTRUM, MOLYBDENUM-
REFLECTED CRITICAL ASSEMBLY

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May, 1970

This information is being published in preliminary form in order to expedite its early release.

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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

E-5641

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INTRODUCTION

A series of critical experiments, which contain materials being considered for a small fast-spectrum reactor, is being performed by Atomics International under contract to Lewis Research Center, contract number NAS 3-12982. The first of the critical assemblies has already been constructed. The completed assembly, which is to be formed in stages, will be a simulation of a conceptual fast reactor which is to be cooled by lithium, fueled with uranium-235 nitride, and reflected by molybdenum (fig. 1).

In the fast reactor concept cylindrical uranium nitride pellets are contained in tantalum alloy (T-111) tubes lined with tungsten foil. A T-111 honeycomb lattice is used to maintain the position of the fuel elements. Six control drums containing both fuel and neutron absorber (T-111) are used for reactivity control. The neutron reflector is made of a molybdenum alloy (TZM).

The initial critical assembly, described in reference 1, had enough void space to permit adding other materials, without changing the overall geometry. Each added material represents some material that may be used in the conceptual reactor. After the addition of each material the fuel loading will be adjusted until the assembly with the new composition becomes critical. Precritical calculations for the initial critical

assembly were reported in reference 1. In that reference the fuel loading requirements and neutron energy spectrum were predicted and the analytical techniques and models used for the calculations were reported.

The present report discusses the effects of lithium nitride, hafnium, tantalum, and tungsten on the flux spectrum and multiplication factor when each is added cumulatively to the critical assembly.

DESCRIPTION OF THE CRITICAL ASSEMBLY

Figure 2 is a sketch of the entire critical assembly; figure 3 shows a cross section of one quadrant of the core portion of this assembly. The outline of the honeycomb tubes depicts the fuel element core lattice formed by grid plates. Similarly grid plates for each control drum locate the positions of fuel elements as shown. The center-to-center spacing (pitch) between tubes in the nondrum region of the core is 0.872 inches (2.215 cm). The entire assembly consists of 247 fuel elements including 11 in each control drum.

A cross section of a fuel element is shown in figure 4. It is shown with a full complement of or alloy fuel (93.2 percent enriched in uranium-235). The space between the fuel bundle and the fuel tube can be used to insert foils of materials proposed for the conceptual fast reactor, such as hafnium, tungsten, and tantalum. The eccentric annular space between the fuel tube and the honeycomb tube can be used for the addition of lithium nitride which represents both the lithium coolant and the nitrogen (in the uranium nitride fuel) in the conceptual reactor. This annular space is eccentric so that the reactivity effects of radial fuel movement can be studied by a series of static critical experiments in which the fuel elements are incrementally rotated.

CALCULATIONAL PROCEDURE

The two-dimensional transport calculations of the critical experiments use the same techniques as described in references 1 to 4. The calculations reported here are based on the latest dimensions and material specifications available.

The GAM-II program (ref. 5) is used to obtain 4-group cross sections; the energy boundaries of the 4 groups are shown in table I. The broad group cross sections for the assembly materials are averaged over three separate spectra which result from fission spectra interacting with a homogeneous core, a molybdenum radial reflector, and a homogeneous axial reflector. It is the composition of the homogeneous core that changes when materials are added (table II).

Two separate spatial calculations are made to estimate the effects of the added materials on the critical mass of oralloy fuel. A two-dimensional (x, y) model can represent the detailed aximuthal asymmetry at the periphery of the core that is shown in figure 3. In this model, the geometry in the direction perpendicular to the (x, y) plane is accounted for by an assigned effective core height H_{eff} , from which the axial neutron leakage is computed (ref. 1). On the other hand, a two-dimensional (r, z) model (fig. 5) treats both the core height and axial reflector regions explicitly, but aximuthal symmetry is imposed. Figure 5 shows the volume fractions for two different loadings of oralloy fuel.

A one-dimensional radial calculation, using the radial dimensions and material composition of the (r, z) model, is used to obtain a consistent value of H_{eff} for the (x, y) model. The effective height is that which provides the same multiplication factor as the radially equivalent (r, z) model calculation.

For the present (x, y) calculations a mesh grid of (39, 35) intervals over the quadrant shown in figure 3 was used. The order of the angular quadrature was S_4P_0 for the (r) and (r, z) models and S_2P_0 and S_4P_0 for the (x, y) model. The TDSN program (ref. 6) was used for the spatial calculations.

An error in the GAM-II compilation of evaluated tantalum inelastic cross sections has been corrected. In order to account for the corrected listing of tantalum cross sections, the multiplication factors of reference 1 must be decreased by 0.0003. (This correction was obtained by an (x, y) calculation.) The present report contains results using corrected Ta cross sections.

DISCUSSION OF RESULTS

The 99-energy group flux spectra obtained from the GAM-II program are shown in figure 6. The spectra result from three homogeneous compositions, each containing 176.61 kilograms of oralloy fuel. The first composition is the core with void space. The second is the core containing 10.695 kilograms of lithium-7 nitride (Li_3^7N), in which each of 247 fuel element tubes contains 43.3 grams of Li_3^7N (98 percent of theoretical density). The third composition is the core with 10.7 kilograms Li_3^7N , 4 kilograms hafnium (Hf), 21 additional kilograms tantalum (Ta), and 40 kilograms tungsten (W).

The effect of the lithium resonance at 250 keV volts is clearly seen in the spectrum of figure 6. Also the overall shift of the spectrum toward lower energies shows the moderating effect of the lithium. The addition of Li_3^7N caused the median energy of the flux to decrease from 0.86 to 0.75 MeV. (Note that ref. 1 reports the median energy as 0.78 MeV; this energy has been corrected to 0.86 MeV.) Table II lists the median energies of the spectra resulting from several core compositions. The addition of 4 kilograms of hafnium to the core already containing Li_3^7N affected the spectrum only slightly and hence this spectrum is not shown in figure 6.

The other spectrum in figure 6 which shows the effect of the Li_3^7N shows that the 4 kilograms Hf, the additional 21 kilograms Ta, and the 40 kilograms W had little effect on the spectrum except to lower the median energy from 0.75 to 0.62 MeV.

Table III lists the multiplication factors from the criticality calculations. The results of three types of calculations are shown; the (r, z) that is necessary to establish the effective height, the $S_2(x, y)$, and the more accurate but longer running $S_4(x, y)$. The multiplication factors in table III are quoted to five significant figures because the spatial calculations are converged to 0.0001. Moreover, some of the effects observed, for example the correction of the tantalum cross sections, require this many significant figures. It should be further noted that the neutron balance, a partial indication of the accuracy of a calculation,

in the three model calculations varied from $+0.2 \times 10^{-5}$ for the $S_4P_0(r, z)$ to -0.5×10^{-3} for the $S_2P_0(x, y)$ to -10^{-3} for the $S_4P_0(x, y)$.

Reference 1 reported a bias between an S_4P_0 -4 group and a more accurate S_8P_1 -13 group approximation of -1.3 percent $\Delta k/k$ which should be removed from the $S_4P_0(x, y)$ multiplication factor. An additional bias of +0.6 percent $\Delta k/k$ was introduced by miscellaneous structural material around the reactor which were not included in the models. The remaining bias of -2.9 percent $\Delta k/k$ was obtained by calculating certain other critical experiments; this large bias is believed to result from relatively poor knowledge of the inelastic down scattering cross sections of tantalum and molybdenum. The total bias in reference 1 then is -3.6 percent $\Delta k/k$.

Exploratory calculations have indicated that decreasing the median energy of the neutron energy spectrum will decrease the bias. And from table II it is evident that the median energy decreases as materials are added cumulatively to the core. Therefore, the decrease in the bias must then be added to the calculated worth of a material to obtain the predicted worth.

However, at the present time we do not have a method for quantitatively correcting the undefined portion of the bias. And since the undefined portion of the bias is the larger part, it will not help the accuracy to do the higher order calculations necessary to correct for the other smaller portions of the bias. Thus, the 3.6 percent bias is assumed to be a constant and it must be removed from the $S_4P_0(x, y)$ calculated multiplication factors given in table III in order to obtain the predicted multiplication factors. However, as a consequence, the Li_3^7N , Hf, Ta, and W worths calculated with this constant bias are only lower limits; the worths should be greater.

The pressure vessel was not included in the (x, y) calculations but its worth (+0.0056) is included in the multiplication factors in table III. The worth of the rubber (neoprene) end pieces of the fuel tubes (illustrated in fig. 7) is +0.0003, but this is not included in the multiplication

factors. Both the pressure vessel and neoprene worths were calculated with the (r, z) model.

In table III case 1 (179.49 kg oralloy) is taken from reference 1 in order to relate the present calculations to those of that reference. This loading (179.49 kg) requires fuel elements which use all seven of the rods and either few or none of the two sizes of fuel wires (fig. 4).

Case 2, with a fuel loading of 176.61 kilograms, was used as the base for the present work. This loading is obtained by using all wires and all rods but the center rod in figure 4.

Case 3 is the 176.61 kilograms loading with 10.7 kilograms of Li_3^7N added uniformly to the core. From the $\text{S}_4\text{P}_0(x, y)$ calculations the total worth of the Li_3^7N is $+0.0023 \Delta k/k$ per kilogram Li_3^7N . Case 4 contains the Li_3^7N and has a smaller fuel loading of 173.01 kilograms, which corresponds to using all but the center rod, all large wires, and no small wires (fig. 4).

From cases 3 and 4 the fuel coefficient with Li_3^7N in the core ($+0.0032 \Delta k/k$ per kg oralloy) is the same as when Li_3^7N is not the core (cases 1 and 2). With this fuel coefficient the reduction in fuel loading brought about by adding the Li_3^7N will be greater than 7.5 kilograms.

Case 5 is the same as case 3 but with 4 kilograms of hafnium added uniformly to the core. The total hafnium worth, in an (x, y) calculation, is $+0.0003 \Delta k/k$. Case 6 has 21 additional kilograms Ta in the core; the total Ta worth is $-0.0007 \Delta k/k$. Case 7 is the same as case 6 but with 40 kilograms W added; the total W worth is $+0.0047 \Delta k/k$.

CONCLUDING REMARKS

This report presents a calculation of the change in fuel loading requirements and the resulting neutron energy spectrum when lithium-7 nitride, hafnium, tantalum, and tungsten are added to a fast spectrum critical assembly. The series of critical experiments is being conducted by Atomics International under contract number NAS 3-12982.

The results of the analysis are as follows:

1. Addition of 10.7 kilograms of Li_3^7N uniformly to the fuel elements of this critical experiment should reduce the critical or alloy loading by more than 7.5 kilograms.
2. The median energy of the neutron energy spectrum should be reduced from 0.86 to about 0.75 MeV by the addition of Li_3^7N . The effect of the lithium resonance should be evident at about 250 keV.
3. Uniform addition of 4 kilograms of natural hafnium to the fuel elements which contain Li_3^7N should be worth about $+0.0003 \Delta k/k$.
4. A negligible change in the spectrum is calculated for the hafnium addition.
5. Addition of Ta has little effect on reactivity; 21 kilograms of Ta is worth about $-0.0007 \Delta k/k$.
6. Addition of 40 kilograms of W should be worth more than $+0.0047 \Delta k/k$.
7. The total effect of the Li_3^7N , Hf, Ta, and W additions on the median energy of the spectrum is to lower it from 0.86 to 0.62 MeV.

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TABLE I. - GAM ENERGY
GROUP BOUNDARIES

Group	E_{lower}^a	U_{lower}^b
1	0.8209 MeV	2.5
2	.1823	4.0
3	40.868 keV	5.5
4	.414 eV	17.0

$$^a E_{\text{upper}} = 14.9 \text{ MeV}$$

$$^b U_{\text{upper}} = -0.4$$

TABLE II. - MEDIAN ENERGY OF THE SPECTRA
OF VARIOUS CORE COMPOSITIONS

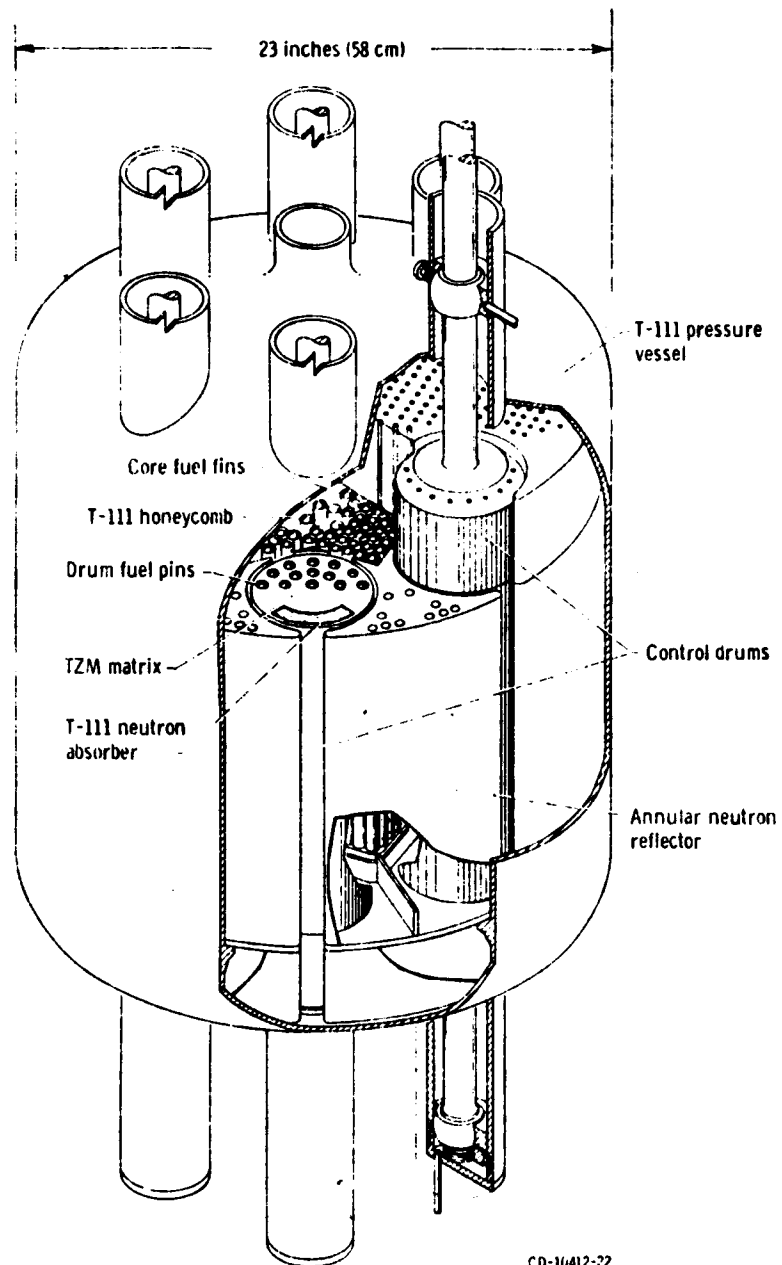
Composition ^a	Median energy, MeV
1 - Core with void	0.86
2 - 10.7 kg Li_3^7N added to composition 1	.75
3 - 4.0 kg Hf added to composition 2	.74
4 - 21.0 kg Ta added to composition 3	.69
5 - 40.0 kg W added to composition 4	.62

^a All compositions contain 176.61 kg of oralloy fuel.

TABLE III. - CALCULATED MULTIPLICATION FACTORS

Case	Composition of homogeneous core	Calculation		
		(r, z)	$S_z(x, y)$	$S_4(x, y)$
1	179.49 kg Oy (ref. 1 - base)		1.0253	1.0354
2	176.61 kg Oy (present - base)	1.0281	1.0159	1.0260
3	Case 2 plus 10.7 kg Li_3^7N	1.0515	1.0430	1.0511
4	173.01 kg Oy +10.7 kg Li_3^7N	1.0392		
5	Case 3 plus 4 kg Hf	1.0517	1.0433	^a 1.0514
6	Case 5 plus 21 kg Ta	1.0501		1.0507
7	Case 6 plus 40 kg W	1.0541		1.0556

^a S_4 multiplication factor estimated by comparison with S_4 (Case 3 and Case 5).



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Figure 1. - Conceptual reactor.

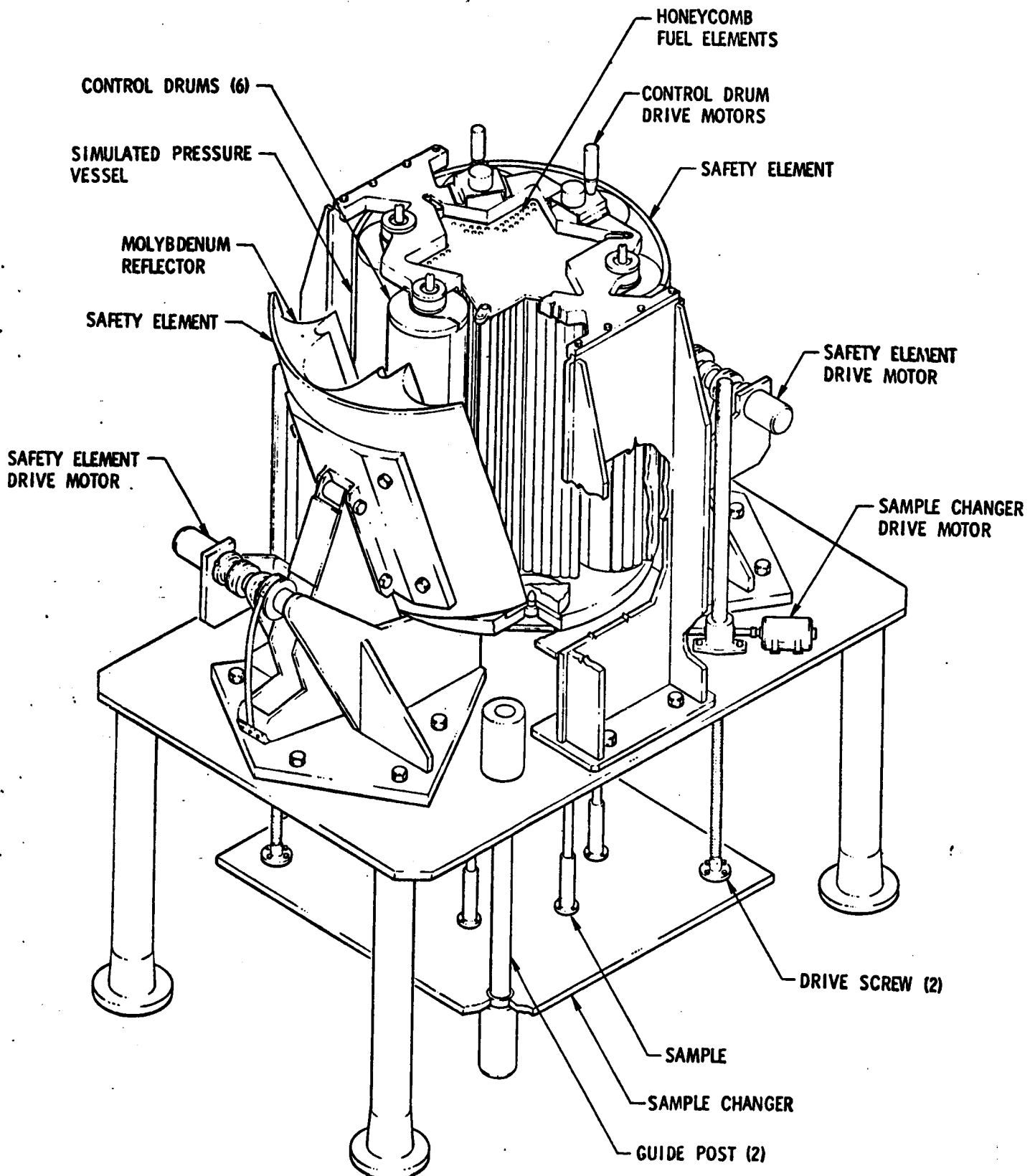


Figure 2. - Critical machine.

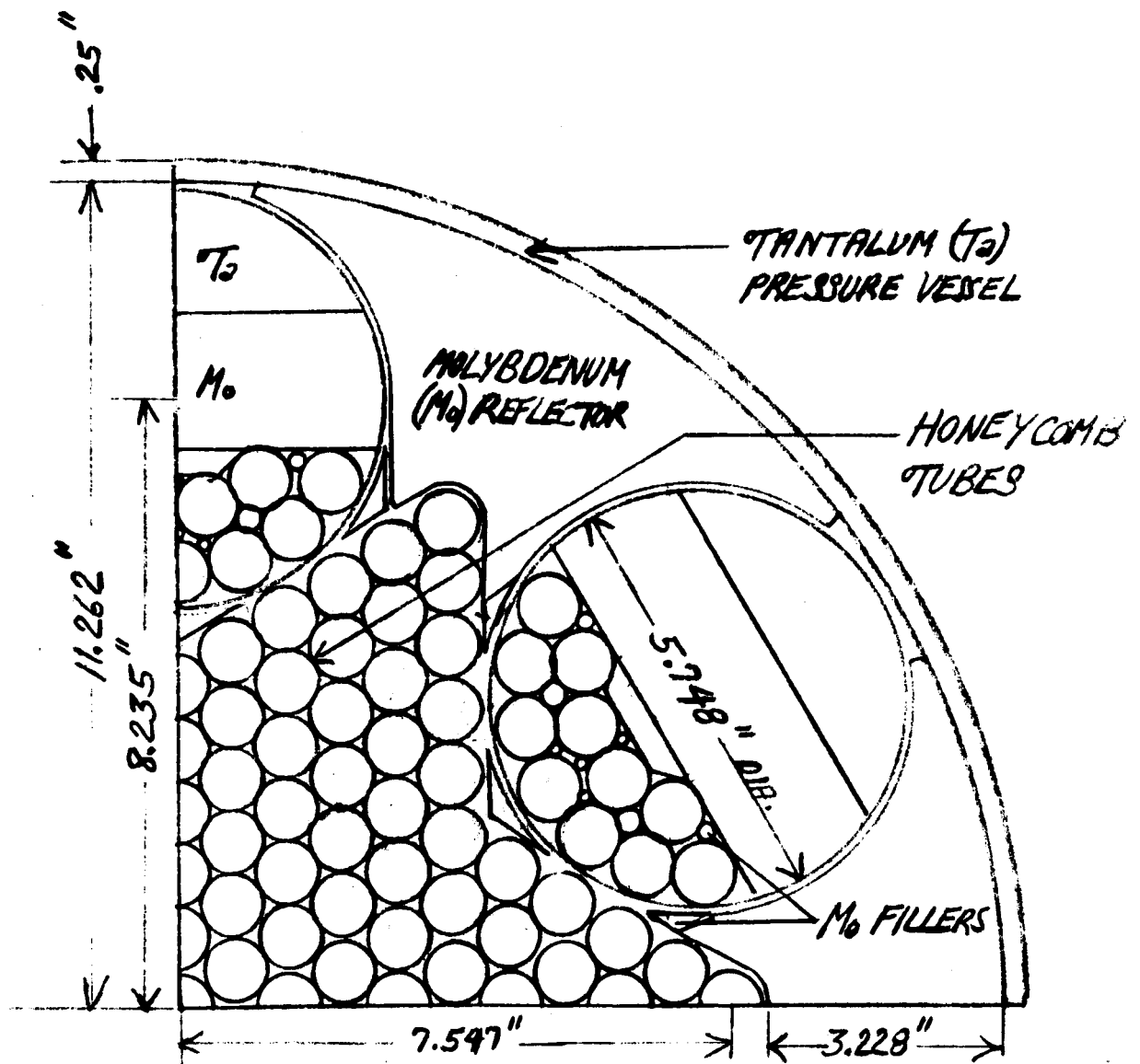


Figure 3. - Cross section of a reactor quadrant.

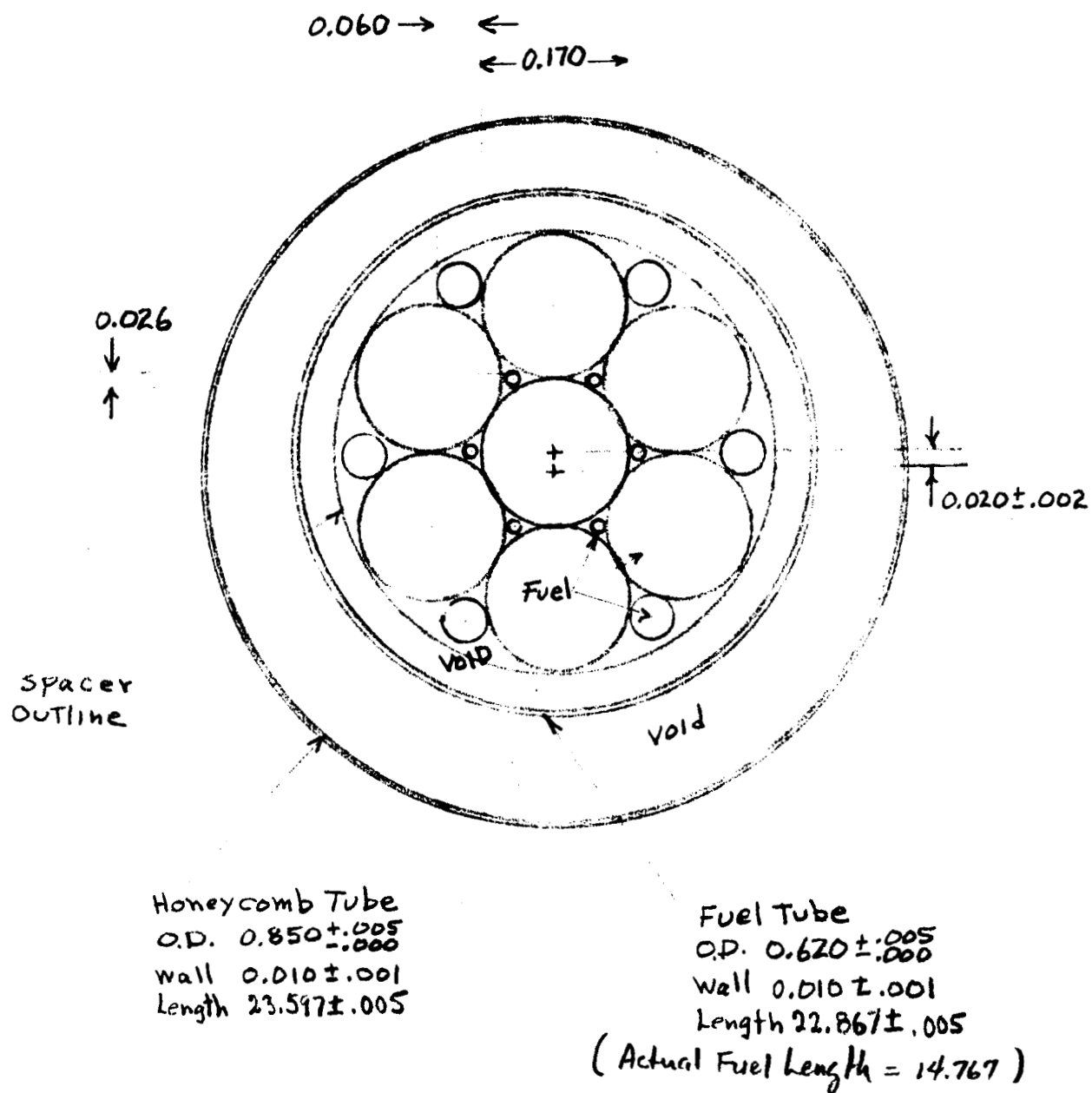


Figure 4. - Fuel element geometry. Dimensions in inches.

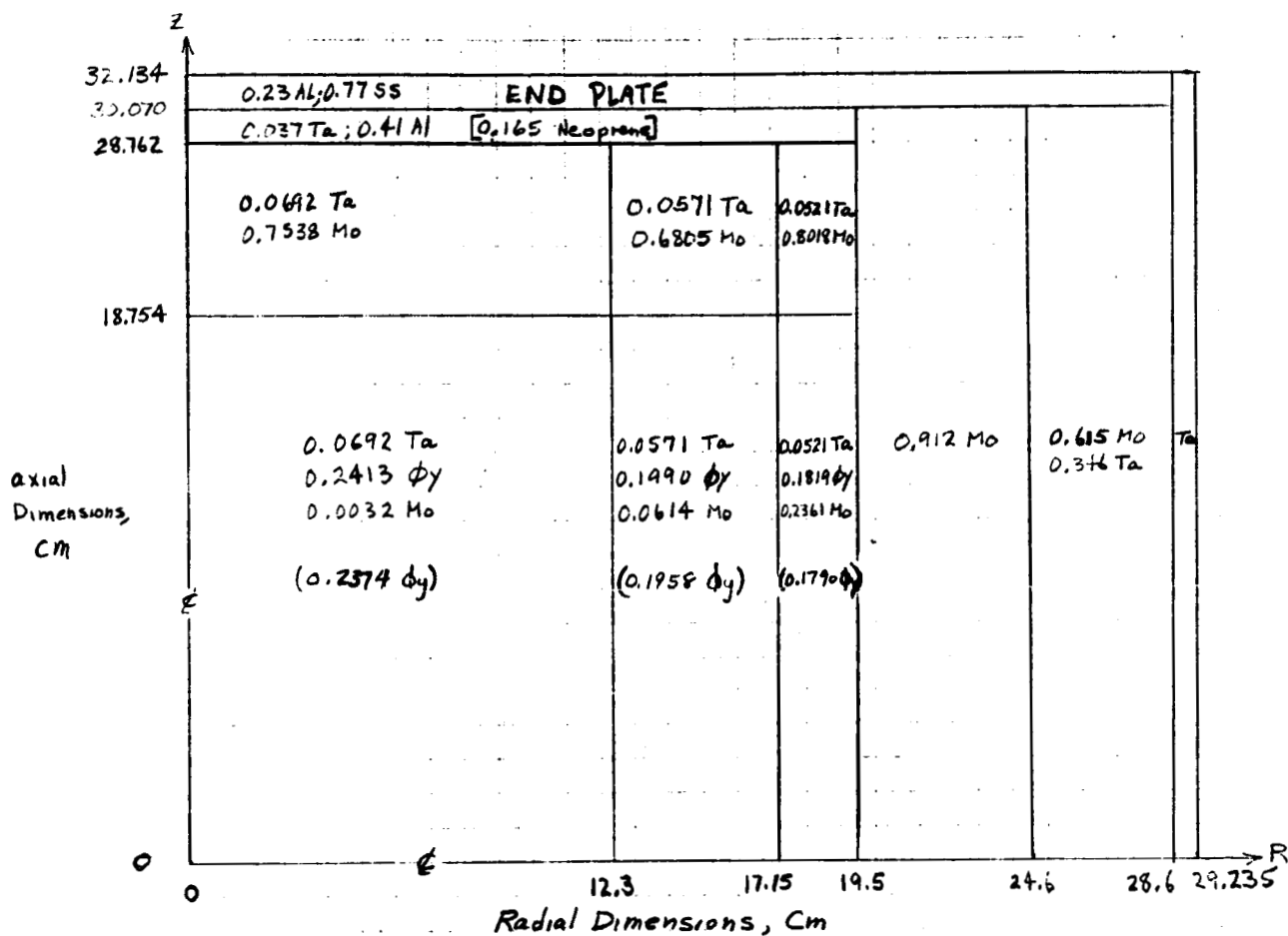


Figure 5. - R,Z calculation geometry showing volume fractions of materials in each region. Fuel loading is 179.49 (176.61) kg of or alloy (ϕy).

$$\int \phi(u) du = 1$$

- (1) CORE ($E_{MED} = 0.86 \text{ MeV}$)
- - (2) CORE WITH Li_7N ($E_{MED} = 0.75 \text{ MeV}$)
- · - (5) CORE WITH $\text{Li}_7\text{N}, \text{HF}, \text{Ta}, \text{W}$ ($E_{MED} = 0.62 \text{ MeV}$)

$\times 10^{-1}$

NEUTRON FLUX PER UNIT LEITHARGY

$\times 10^{-3}$

MEDIAN ENERGY
(X295)
0.86 0.75 0.62
MeV

$\times 10^{-4}$

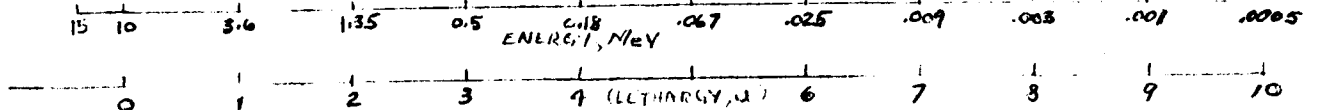


Figure 6. - GAM-II core flux spectrum.

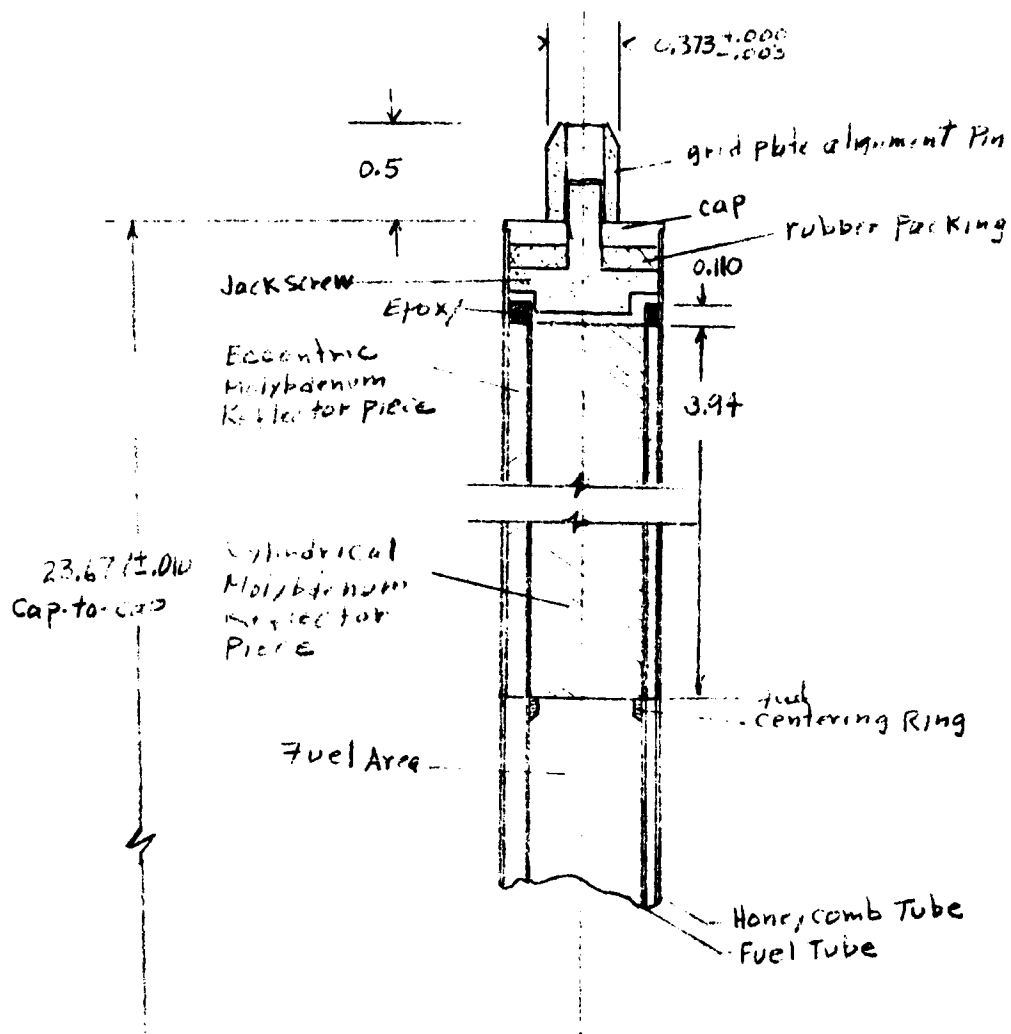


Figure 7. - Fuel element closure method and end reflector design. All dimensions in inches.